

# A Bayesian approach to magnetic moment determination using $\mu$ SR

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## Abstract

A significant challenge in zero-field  $\mu$ SR experiments arises from the uncertainty in the muon site. It is possible to calculate the dipole field (and hence precession frequency  $\nu$ ) at any particular site given the magnetic moment  $\mu$  and magnetic structure. One can also evaluate  $f(\nu)$ , the probability distribution function of  $\nu$  assuming that the muon site can be anywhere within the unit cell with equal probability, excluding physically forbidden sites. Since  $\nu$  is obtained from experiment, what we would like to know is  $g(\mu|\nu)$ , the probability density function of  $\mu$  given the observed  $\nu$ . This can be obtained from our calculated  $f(\nu/\mu)$  using Bayes' theorem. We describe an approach to this problem which we have used to extract information about real systems including a low-moment osmate compound, a family of molecular magnets, and an iron-arsenide compound.

**Keywords:** Muon-spin rotation, Dipole fields, Muon sites

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## 1. Introduction

In a  $\mu$ SR experiment the muon-spin precession frequency  $\nu$  can be used to deduce the local magnetic field  $B$  at the muon site. If there are a number of muon sites with different local fields  $\{B_j\}$ , then the  $\mu$ SR signal can contain a number of components with frequencies  $\{\nu_j\}$ . In complex systems it can be highly non-trivial to determine the precise location of the muon site or sites. However, it is nevertheless useful to attempt to extract an estimate of the magnitude of the moment of the magnetic species producing the local field, even in the face of ignorance of the location of the muon site or sites. In this paper we describe a method to attempt this using Bayesian inference. The paper is structured as follows: in Sections 2 and 3 we review the theory of dipolar fields and Bayes' theorem respectively to provide the necessary background to the calculation which is described in Section 4.

## 2. Dipolar fields

An implanted muon spin precesses around a local magnetic field,  $B_{\text{local}}$ , with a frequency  $\nu = (\gamma_\mu/2\pi)|B_{\text{local}}|$ , where  $\gamma_\mu/2\pi = 135.5 \text{ MHz T}^{-1}$ . The magnetic field  $B_{\text{local}}$  at the muon site is given by

$$B_{\text{local}} = B_0 + B_{\text{dipole}} + B_L + B_{\text{demag}} + B_{\text{hyperfine}}, \quad (1)$$

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where  $\mathbf{B}_0$  represents the applied field (zero in the experiments considered here),  $\mathbf{B}_{\text{dip}}$  is the dipolar field from magnetic ions,  $\mathbf{B}_L = \mu_0 \mathbf{M}/3$  is the Lorentz field,  $\mathbf{B}_{\text{demag}}$  is the demagnetizing field from the sample surface and  $\mathbf{B}_{\text{hyperfine}}$  is the contact hyperfine field caused by any spin density overlapping with the muon wavefunction. In antiferromagnets the Lorentz and demagnetizing fields vanish (in polycrystalline ferromagnets they cancel to some extent). The contact hyperfine field is hard to estimate but we will neglect it. The remaining term is the dipolar field  $\mathbf{B}_{\text{dip}}$  and is a function of the muon-site  $\mathbf{r}_\mu$ . It can be written as

$$\mathbf{B}_{\text{dip}}^\alpha(\mathbf{r}_\mu) = \sum_i D_i^{\alpha\beta}(\mathbf{r}_\mu) m_i^\beta, \quad (2)$$

a sum over the magnetic ions; the magnetic moment of the  $i$ th ion is  $\mathbf{m}_i$ . In Eq. (2),  $D_i^{\alpha\beta}(\mathbf{r}_\mu)$  is the dipolar tensor given by

$$D_i^{\alpha\beta}(\mathbf{r}_\mu) = \frac{\mu_0}{4\pi R_i^3} \left( \frac{3R_i^\alpha R_i^\beta}{R_i^2} - \delta^{\alpha\beta} \right), \quad (3)$$

where  $\mathbf{R}_i \equiv (R_i^x, R_i^y, R_i^z) = \mathbf{r}_\mu - \mathbf{r}_i$  and  $\delta^{\alpha\beta}$  is the Kronecker delta ( $\delta^{\alpha\beta} = 1$  if  $\alpha = \beta$ , else  $\delta^{\alpha\beta} = 0$ ). The behaviour of this tensor is dominated by the arrangement of the nearest-neighbour magnetic ions and leads to a non-zero local magnetic field for almost all possible muon sites, even in an antiferromagnetically ordered system [1, 2]. The sum in Eq. (2) is taken over the infinite lattice, but it is well known [3] that this sum converges in such a way that it is necessary only to sum over points inside a sphere centred on  $\mathbf{r}_\mu$  with sufficiently large radius. An alternative method of calculation is provided by the method of Ewald summation (for details see [4]).

### 3. Bayes' theorem

We recap some elementary probability theory [5, 6]. The *conditional probability*  $P(A|B)$  is the probability that event A occurs *given* that event B has happened. The *joint probability*  $P(A \cap B)$  is the probability that event A and event B both occur. The joint probability  $P(A \cap B)$  is equal to the probability that event B occurred multiplied by the probability that A occurred, given that B did, i.e.,

$$P(A \cap B) = P(A|B)P(B), \quad (4)$$

and, equally well,

$$P(A \cap B) = P(B|A)P(A). \quad (5)$$

Now consider the case where there are a number of mutually exclusive events  $A_i$  such that

$$\sum_i P(A_i) = 1. \quad (6)$$

Then we can write the probability of some other event X as

$$P(X) = \sum_i P(X|A_i)P(A_i). \quad (7)$$

In very general terms, one can say that given some hypothesis H there usually exists some computational strategy to evaluate the probability of a particular outcome O assuming that hypothesis to be correct (i.e., there is some method to compute the quantity  $P(O|H)$ ). However, what you often want to do is the reverse of this: you know the outcome because it has actually occurred and you want to choose an explanation out of the possible hypotheses. In other words, given the outcome you want to know the probability that the hypothesis is true, and the problem is that  $P(H|O)$  is typically much more challenging to evaluate. The needed transformation of  $P(O|H)$  into  $P(H|O)$  can be accomplished using *Bayes' theorem* (named after Thomas Bayes (1702–1761), although the modern form is due to Laplace). This theorem can be stated as follows:

$$P(A|B) = \frac{P(A)P(B|A)}{P(B)}. \quad (8)$$

Here  $P(A)$  is called the *prior probability*, since it is the probability of A occurring without any knowledge as to the outcome of B. The quantity which you derive is  $P(A|B)$ , the *posterior probability*. The proof of Bayes' theorem is

very simple: one simply equates Eqs. (4) and (5) and rearranges. For the purpose of this paper, we will write Bayes' theorem using Eq. (7) as

$$P(\mu|\nu) = \frac{P(\mu)P(\nu|\mu)}{\int P(\nu|\mu')P(\mu') d\mu'}. \quad (9)$$

#### 4. Calculation

Since the positive muon seeks out areas of negative charge density, constraints can be placed on the likely location of stopped muons. For example, the muon is unlikely to stop close to the positively charged ions in a system. In many oxides, muons have been shown to stop around 0.1 nm from an  $O^{2-}$  ion [7]. In our calculations, we assume a magnetic moment  $\mu$  on the magnetic species in our material and consider a particular magnetic structure. Positions in the unit cell are then generated at random and, provided the relevant constraints are satisfied, the dipole field is calculated at each of them. After many such randomly generated positions, one obtains a distribution of dipole fields. (This distribution sometimes have sharp features associated with them which are van Hove singularities [2, 8].) The magnitudes of the resulting dipole fields are then converted into muon precession frequencies, and the resulting histogram yields the probability density function (pdf)  $f(\nu/\mu)$ , evaluated as a function of precession frequency  $\nu$  divided by magnetic moment  $\mu$  (since the precession frequency scales with the magnetic moment). This function  $f(\nu/\mu)$  allows us to evaluate  $P(\nu|\mu)$  of Eq. (9). We can write

$$P(\nu|\mu) = \frac{1}{\mu} f(\nu/\mu). \quad (10)$$

The function  $f(\nu/\mu)$  is normalized so that  $\int f(\nu/\mu) d(\nu/\mu) = 1$ , and hence the factor of  $\frac{1}{\mu}$  is needed in Eq. (10) so that  $\int P(\nu|\mu) d\nu = 1$ . An example of this approach is shown in Fig. 1(a) for a cubic lattice of antiferromagnetically coupled spins. The solid line shows the case when a simple constraint is applied so that the muon is not permitted to stop at a site closer than a particular critical distance to the magnetic moments, resulting in a cut-off of the tail at high frequency.

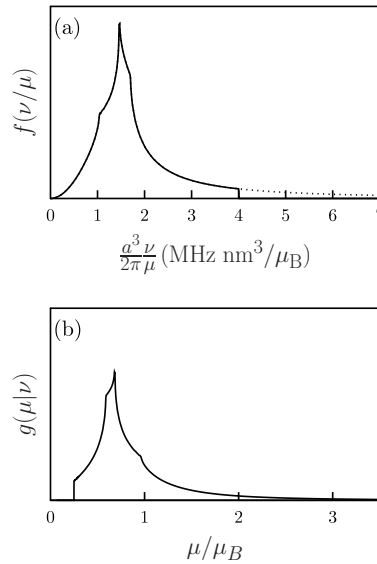


Figure 1: (a) The dipolar field distribution for a simple cubic lattice of antiferromagnetically aligned magnetic moments parallel to [110] (with antiferromagnetic wave vector  $\mathbf{q} = (\pi, \pi, \pi)$ ). The lattice parameter is  $a$  and the size of the moment is  $\mu$  [2]. The solid (dashed) line shows the distribution with a cut-off (not) applied. (b) The extracted pdf for the moment given a particular observed frequency ( $\nu = 2\pi\mu_B/a^3$ ).

Since  $\nu$  is obtained from a real experiment, what we would like to know is  $g(\mu|\nu)$ , the pdf of  $\mu$  given the observed  $\nu$ . This can be obtained from our calculated  $f(\nu/\mu)$  using Bayes' theorem in the form of Eq. (9), which yields

$$g(\mu|\nu) = \frac{\frac{1}{\mu} f(\nu/\mu)}{\int_0^{\mu_{\max}} \frac{1}{\mu'} f(\nu/\mu') d\mu'}, \quad (11)$$

where we have assumed a prior probability  $[P(\mu)]$  for the magnetic moment that is uniform between zero and  $\mu_{\max}$ , and so  $P(\mu)$  is replaced by the uniform probability density  $1/\mu_{\max}$  [which cancels on the top and bottom of Eq. (11)]. We choose  $\mu_{\max}$  to take a large value, although we have found that our results are insensitive to the precise value of  $\mu_{\max}$ . A very simple example of this approach is shown in Fig. 1(b). When multiple frequencies  $\nu_i$  are present in the spectra, it is necessary to multiply their probabilities of observation in order to obtain the chance of their simultaneous observation, so we evaluate  $g(\mu|\{\nu_i\}) \propto \prod_i \int_{\nu_i - \Delta\nu_i}^{\nu_i + \Delta\nu_i} f(\nu_i/\mu) d\nu_i$ , where  $\Delta\nu_i$  is the error on the fitted frequency.

We have now applied this technique to  $\mu$ SR data a variety of real systems in which the muon site is not known. These include  $\text{Ba}_2\text{NaOsO}_6$  in which we can show from the observed precession frequencies that the magnetic ground state is most likely to be low-moment ( $\approx 0.2\mu_B$ ) ferromagnetism and not canted antiferromagnetism [9]. We have also used it to show a reduced moment in the two-dimensional molecular magnet  $[\text{Cu}(\text{HF}_2)(\text{pyz})_2]\text{BF}_4$  [10] and in the pnictide superconductor  $\text{NaFeAs}$  [11]. In all these cases we do not have a priori information concerning the muon site but can nevertheless place bounds upon the magnetic moment from the observed precession signal using this technique. A possible drawback that should be borne in mind is that the hyperfine contribution to the local field is neglected and if this is significant it could affect the conclusions drawn. As many of the systems examined so far using this technique have localized, reduced moments and lower-frequency precession signals, it is probable that the hyperfine contribution is not significant in these cases.

## 5. Acknowledgments

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